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## Optically induced spin disorder in $\text{YVO}_3$

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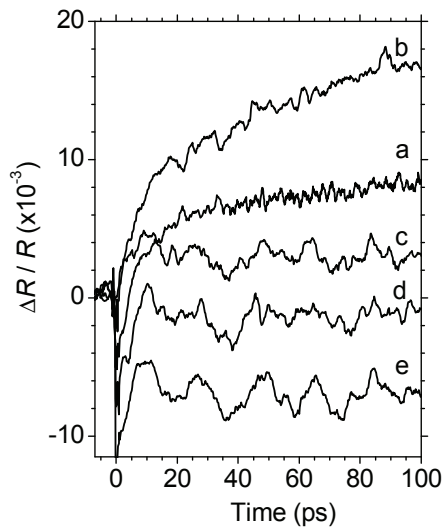
**Abstract.** Yttrium vanadate ( $\text{YVO}_3$ ) is an interesting material for studying optically induced phase transitions because it exhibits a rich variety of coupled structural and correlated electronic phases. We show that optical excitation effectively decouples spin and magnetic order and results in rapid spin melting within 4 ps while orbital order survives for longer than 100 ps.

Solid to solid phase transitions play a key role in modern optical rewritable data storages like CDs and DVDs. The switching rate of such optical memory devices is ultimately limited by the phase-change rate. Present-day memory devices, however, operate under thermodynamic conditions, and their performance is limited by heat transport to the substrate. Improving memory devices poses a fundamental problem and requires an extensive knowledge on the nature and dynamics of optically induced phase transitions, in particular of non-thermal transitions. Such ultrafast solid-solid phase transitions may go already on pico- and even subpicosecond timescales, much faster than phonon-phonon or even electron-phonon equilibration [1]. Today perovskite materials gain considerable interest due to their advantages in fast manipulation of strongly coupled magnetic and orbital order, spontaneous electric polarization, and /or ferroelasticity. In such materials rapid charge [2], orbital [3] and spin transitions [4] has been reported. In this paper we study optically induced phase transitions in  $\text{YVO}_3$ , which possesses coupled spin and orbital ordering below Néel temperature.

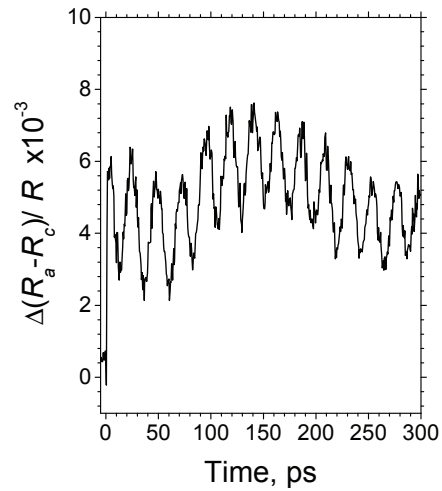
The perovskite  $\text{YVO}_3$  possesses a sequence of orbital and spin transitions as a function of temperature. At room temperature this crystal is paramagnetic and has an orthorhombic  $Pbnm$  structure. Below  $T_{\text{OO}}=200$  K the  $d$ -orbitals form a  $G$ -type ordering structure, in which the orbital occupation periodically alternates in all three crystallographic directions [5,6]. This transition is accompanied by a symmetry lowering into a  $P2_1/b11$  monoclinic form [7]. Another phase transition at  $T_{\text{N}} = 116$  K is associated with  $C$ -type spin ordering forming an antiferromagnetic arrangement in the  $ab$  plane with a ferromagnetic arrangement along the  $c$  axis. Finally, at  $T_{\text{CG}} = 77$  K,  $\text{YVO}_3$  undergoes a first-order phase transition, at which spin and orbital orderings change from  $C$ - to  $G$ -type and from  $G$ - to  $C$ -type spin, respectively. This phase transition is accompanied by an unusual increase in the symmetry of the lattice back to the orthorhombic  $Pbnm$  form [6-9]. It is known that weak interaction of neighbouring  $d$  orbitals with light forms three spectral bands located at 1.8 eV, 2.4 eV, and 3.3 eV. The optical strength of these bands strongly depends on the spin and orbital arrangement [7]. As a result, each phase of  $\text{YVO}_3$  possesses a unique fingerprint in the optical absorption and reflection spectra and thus allows the dynamics of the phase transitions to be traced by monitoring the optical reflectivity.  $\text{YVO}_3$  has a phase diagram similar to  $\text{LaVO}_3$ , for which nonthermal melting of the orbital order was recently reported [3]. However, an important difference is that spin and orbital order phase

transition temperatures in  $\text{LaVO}_3$  are so close to each other that the different phases are not distinguishable in optically induced experiments. Fortunately, the transitions in  $\text{YVO}_3$  are far apart allowing for a study multiple phase changes.

Time-resolved two-colour pump-probe experiments have been carried out on single crystal  $bc$ -oriented polished platelets of an  $\text{YVO}_3$ . Details of the sample growth can be found in reference [7]. The sample has been kept in helium-flow cryostat where it was excited by an optical pump pulse train with a repetition rate of 500 Hz. The dynamics of photoinduced changes were monitored by weak probe pulses focused onto the sample surface in the central area of the pump excitation.

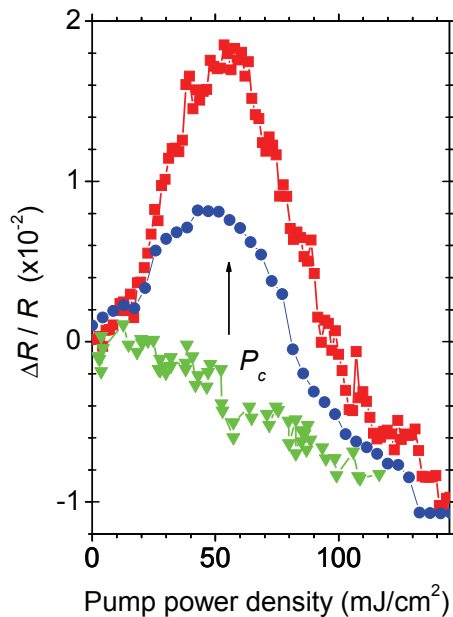


**Figure 1** Kinetics of transient reflectivity probed by 630 nm light after optical excitation with 800 nm and power density of *a*: 25 mJ/cm<sup>2</sup>, *b*: 45 mJ/cm<sup>2</sup>, *c*: 85 mJ/cm<sup>2</sup>, *d*: 100~mJ/cm<sup>2</sup>, and *e*: 170 mJ/cm<sup>2</sup>.

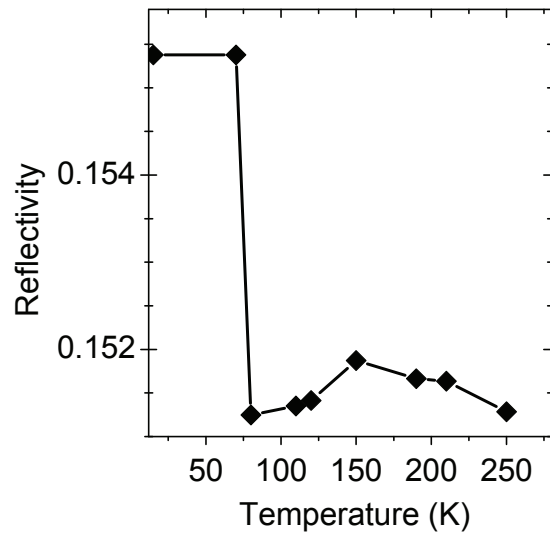


**Figure 2** Transient anisotropy of the  $ac$  plane reflectivity probed by 800 nm light and pumped by 400 nm.

Figure 1 demonstrates time traces of the transient reflectivity for different excitation power densities measured at 25 K, *i.e.* in the lowest temperature phase. For all excitation power densities the transient reflectivity shows an abrupt negative change just after arrival of the pump pulse. This rapid change is followed by pronounced temporal oscillations in the transient reflectivity amplitude with a period of about 19 ps. These oscillations are a result of the interference of the probe light reflected on the sample surface and on the acoustic strain wave formed on the sample surface by the optically induced stress [10]. This acoustic wave propagates in a direction perpendicular to the surface with the velocity  $s = \lambda_{probe} / 2n\tau_{os}$ , with  $n$  the refractive index of  $\text{YVO}_3$ ,  $\lambda_{probe}$  the wavelength of the probe light, and  $\tau_{os}$  the oscillation frequency in the reflectivity. Using our experimental data  $\tau_{os} = 19$  ps taken from figure 1 and knowing [7]  $n=2.3$  at  $\lambda_{probe}=630$  nm we are able to estimate  $s = 7.5 \pm 0.5$  km/s, which is close to the longitudinal sound velocity measured in another vanadate perovskite [11]. These oscillations are more vivid in the trace of transient difference in reflectivity along the  $a$  and  $c$  axes, the induced anisotropy (figure 2). Here, the probe wavelength was set to 800 nm resulting in a proportional increase in the period of oscillations up to 23 ps. The nonoscillating part of the transient reflectivity shown in figure 1 can be quite faithfully fitted by a double-exponential decay function with  $\tau_1 = 3$  ps and  $\tau_2 = 45$  ps for the fast and slow decay terms, respectively.



**Figure 3** Power dependence of the transient reflectivity probed at 100 ps after optical excitation at 25 K (red squares), 100 K (blue circles), and 140 K (green triangles).



**Figure 4** Stationary reflectance calculated using the optical constants taken from reference [7].

For weak optical excitations (curves *a* and *b* in figure 1) the slow decay dominates the transient reflectivity dynamics; the transient reflectivity is positive over most of the experimental time scale and rises with increasing excitation power density  $P$ . However, when  $P$  is above a critical value the slow decay component suddenly disappears (curves *a* and *b* in figure 1). Further, the saturation level of the transient reflectivity now becomes more negative as the excitation strength increases.

Figure 3 shows the dependence of the transient reflectivity on excitation power density at 100 ps delay for the three initial phases of  $\text{YVO}_3$ : the *C*-type orbital ordered phase at 25 K (red squares), the *G*-type orbital ordered phase with *C*-type spin order at 100 K (blue circles), and the *G*-type orbital ordered phase with disordered spins at 140 K (green triangles). A photoinduced phase transition manifests itself as an anomaly at a critical power  $P_c = 55 \text{ mJ/cm}^2$ , which appears below  $T_N$  only. The observed threshold behaviour provides direct evidence for a photoinduced phase transition. Quite importantly, the shape and the threshold of the power dependence do not change when the sample temperature crosses  $T_{CG}$  (red squares and green triangles). However, when the temperature approaches  $T_N$  from below, the anomaly in the power dependence diminishes (blue circles) and finally disappears at  $T_N$ . For  $T > T_N$  the transient reflectivity has a monotonic linear dependence on the excitation power density (green triangles).

The disappearance of the threshold power behaviour above  $T_N$  indicates that the observed photoinduced phase transition is related to the melting of the spin order. It is interesting to note that melting of spin order for the initial  $T_{CG} < T < T_N$  is similar to a thermal heating of  $\text{YVO}_3$  over  $T_N$ . Indeed, both the variation of the reflectivity with increasing temperature (figure 4) and the induced changes in reflectivity 100 ps after the optical excitation (figure 3, blue circles) are positive in this temperature range. In turn, for  $T < T_{CG}$  the optically and thermally induced changes in the reflectivity are different: Optically induced changes are positive and has maximum at  $P_c$  indicating the occurrence of spin melting. At the same time, when the temperature crosses  $T_{CG}$  from below, the transition from *C*- to *G*-type orbital state is accompanied by an abrupt decrease in reflectivity (figure 4). This indicates that the spin-orbital reorientation transition does not occur even at the excitation powers for which

spin melting takes place, and for  $T < T_{CG}$  the arrangement of spins and orbitals are far from thermal equilibrium even 100 ps after the optical excitation.

Our experimental data suggest the following model. The pump pulse induces an electronic transition from the oxygen  $2p$  band to the empty states in the vanadium  $3d$  band [7]. Subsequently, these electrons relax to lower energy states and their excess energy excites the spin and orbital degrees of freedom. For  $T > T_N$  the change in the reflectivity of  $YVO_3$  is negative and governed by the arrangement of the occupied orbitals, which thermalizes within  $\tau_1$ . In addition, at  $T < T_N$ , the photoexcited hot electrons may also transfer their energy to the ordered spin network, leading to a positive change in the reflectivity. At  $P < P_c$ , the subsequent thermalization of spins with the orbital degree of freedom manifests itself as the slow  $\tau_2$  component in the transient reflectivity traces. The increased spin temperature and excited orbitals thus influence the reflectivity in opposite directions. At  $P > P_c$  the power density is enough to excite the spin network above its melting point, which leads to an anomaly in the power dependency of the transient reflectivity.

The dynamics of the spin-orbital reordering transition is different. At  $T < T_{CG}$  the arrangement of the orbitals remains far from equilibrium even 100 ps after the optical excitation, while excitation power is enough to induce melting of the spin order. This allows us to conclude that the  $C \rightarrow G$  orbital transition does not occur on a picosecond time scale and that  $YVO_3$  undergoes a transition to a metastable phase with disordered spins but  $C$ -ordered orbitals. We note that in our experiment the excitation power density was not sufficient to promote complete orbital melting, but nevertheless may result in partial orbital disorder.

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